Monoclinic phase transformation and mechanical durability of zirconia ceramic after fatigue and autoclave aging

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Abstract: OBJECTIVES This study evaluated the influence of two aging procedures on the biaxial flexural strength of yttria-stabilized tetragonal zirconia ceramics. MATERIAL AND METHODS Disc-shaped zirconia specimens and (ZE: E.max ZirCAD, Ivoclar; ZT: Zirkon Translucent, Zirkonzahn) (N = 80) (∅:12 mm; thickness:1.2 mm, ISO 6872) were prepared and randomly divided into four groups (n = 10 per group) according to the aging procedures: C: Control, no aging; M: mechanical cycling (2 × 106 cycles/3.8 Hz/200 N); AUT: Aging in autoclave at 134℃, 2 bar for 24 h; AUT + M: Autoclave aging followed by mechanical cycling. After aging, the transformed monoclinic zirconia (%) were evaluated using X-ray diffraction and surface roughness was measured using atomic force microscopy. The average grain size was measured by scanning electron microscopy and the specimens were submitted to biaxial flexural strength testing (1 mm/min, 1000 kgf in water). Data (MPa) were statistically analyzed using 2-way analysis of variance and Tukey's test (α = 0.05). RESULTS Aging procedures significantly affected (p = 0.000) the flexural strength data but the effect of zirconia type was not significant (p = 0.657). AUTZT (936.4 ± 120.9b) and AUT + MZE (867.2 ± 49.3b) groups presented significantly higher values (p < 0.05) of flexural strength than those of the control groups (CZT : 716.5 ± 185.7a ; CZE : 779.9 ± 114a) (Tukey's test). The monoclinic phase percentage (%) was higher for AUTZE (71), AUTZT (66), AUT + MZE (71), and AUT + MZM (66) compared to the C groups (ZE:0; ZT:0). Surface roughness (µm) was higher for AUTZE (0.09), AUTZT (0.08), AUT + MZE (0.09 µm), and AUT + MZT (0.09 µm) than those of other groups. CONCLUSIONS Regardless of the zirconia type, autoclave aging alone or with mechanical aging increased the flexure strength but also induced higher transformation from tetragonal to monoclinic phase in both zirconia materials tested. © 2016 Wiley Periodicals, Inc. J Biomed Mater Res Part B: Appl Biomater, 105B: 1972-1977, 2017.

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Monoclinic phase transformation and mechanical durability of zirconia ceramic after fatigue and autoclave aging

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Short title: Mechanical durability of zirconia after aging.

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Abstract

Objectives. This study evaluated the influence of two aging procedures on the biaxial flexural strength of yttria-stabilized tetragonal zirconia ceramics. ***Material and methods.*** Disc-shaped zirconia specimens and (ZE: E.max ZirCAD, Ivoclar; ZT: Zirkon Translucent, Zirkonzahn) (N=80) (∅:12 mm; thickness:1.2 mm, ISO 6872) were prepared and randomly divided into four groups (n=10 per group) according to the aging procedures: C: Control, no aging; M: mechanical cycling ($2x10^6$ cycles/3.8 Hz/200 N); AUT: Aging in autoclave at 134°C, 2 bar for 24 h; AUT+M: Autoclave aging followed by mechanical cycling. After aging, the transformed monoclinic zirconia (%) were evaluated using X-ray diffraction (XRD) and surface roughness was measured using atomic force microscopy (AFM). The average grain size was measured by scanning electron microscopy (SEM) and the specimens were submitted to biaxial flexural strength testing (1 mm/min, 1000 kgf in water). Data (MPa) were statistically analyzed using 2-way ANOVA and Tukey`s test (α=0.05). ***Results.*** Aging procedures significantly affected (p=0.000) the flexural strength data but the effect of zirconia type was not significant (p=0.657). AUT_{ZT} (936.4±120.9[b]) and $AUT+M_{ZE}$ (867.2±49.3[b]) groups presented significantly higher values ($p<0.05$) of flexural strength than those of the control groups (C_{ZT}: 716.5±185.7[a]; C_{ZE}: 779.9±114[a]). The monoclinic phase percentage (%) was higher for AUT_{ZE} (71), AUT_{ZT} (66), $AUT+M_{ZE}$ (71) and $AUT+M_{ZM}$ (66) compared to the C groups (ZE:0; ZT:0). Surface roughness (µm) was higher for AUT_{ZE} (0.09), AUT_{ZT} (0.08), $AUT+M_{ZE}$ (0.09 µm) and $AUT+M_{ZT}$ (0.09 µm) than those of other groups. ***Conclusions.*** Regardless of the zirconia type, autoclave aging alone or with mechanical aging increased the flexure strength but

also induced higher transformation from tetragonal to monoclinic phase in both zirconia materials tested.

Keywords: Aging, Flexural strength, Mechanical cycling, Phase transformation, Yttria-stabilized tetragonal zirconia

Introduction

Yttria-stabilized tetragonal zirconia (hereafter: zirconia) is a polymorphic material that exists in nature in three main forms, namely monoclinic, tetragonal and cubic.[3,10,23] The tetragonal structure may be stabilized at room temperature with the addition of oxides, such as calcium, magnesium, cerium or yttrium, the last being used in dental applications.[17,23,30] Structural transformation from tetragonal into monoclinic occurs under localized stress,[14,17] that is associated with 3-4% volume expansion in zirconia. This induces tension under compression and microcracking around the transformed particles.[3,26] Transformation mechanism works as a barrier making crack propagation difficult thereby increasing the fracture resistance of zirconia.[19,22,29]

Transformation from tetragonal into monoclinic phase may also be induced at low temperature in the humid environment of the oral cavity. This phenomenon is referred to as aging of zirconia due to low-temperature degradation (LTD) (120-400˚C)[3] where water breaks zirconia molecules (ZrO_2), forming zirconia oxide or breaks yttrium molecule into yttrium hydroxide (Y_2O_3).[4] Such transformation is initiated first in isolated surface ceramic grains which then gradually spread along the surface and penetrate into the deep portions of the material in a process called “nucleation and growth”.[1] Despite being very slow at oral temperatures, this degradation mechanism may drastically decrease the mechanical properties of zirconia. Consequently, resistance and tenacity decrease, superficial roughness increase, surface grains detach from the material yielding to microcracks,[9] which may all reduce the long-term success of zirconia implant abutments and other zirconia-based fixed dental prosthesis (FDP).[3,5,17,30] Moreover, several clinical studies reported high incidence of chipping of the veneering ceramic as one of the main failures in zirconia FDPs.[8,24,25] Hence, the use of monolithical zirconia, meaning without veneering ceramic, is increasing for dental applications although it is susceptible to degradation.[11,15]

Aging protocols applied to study LTD of zirconia consists either mechanical cyclic aging[7] or aging in autoclave only.[2] The objectives of this study therefore were to evaluate the flexural strength, surface roughness and transformation change in two types of zirconia after hydrothermal aging and mechanical fatigue. The hypotheses tested were that the aging procedures would decrease the flexural strength and increase the amount of monoclinic phase after all aging procedures.

Materials and Methods

Specimen preparation

Disk-shaped zirconia (ZE: E.max ZirCad, Ivoclar, Vivadent, Schaan, Liechtenstein-ZE and ZT: Zirkon Translucent: Zirkonzahn GmbH, Gais, Italy) specimens were sectioned with a diamond disc (Extec, Enfield, CT, USA) in a cutting machine (Isomet®1000 Precision Saw, Buehler, Lake Bluff, IL, USA) under water irrigation. Specimens were obtained at dimensions suggested by ISO 6872 (∅=15 mm; thickness: 1.6 mm) and sintered. They were polished with 180, 600 and 1200-grit silicon carbide papers in sequence for 5 min each under water-cooling and ultrasonically cleaned in isopropyl alcohol for 8 min. ZE (Zyrcomat T furnace at 1530°C for 120 min) and ZT (Zirkonofen 600/V2 furnace at 1540°C for 120 min) specimens were sintered accordingly to the final dimension of a diameter of 12 mm and thickness of 12 mm. The zirconia discs were not polished before sintering.

Aging procedures

Zirconia specimens (N=80) were randomly divided into four groups (n=10 per group) according to the aging procedures:

C: Control, no aging was performed in this group.

M: Specimens were exposed to mechanical cycling for $2x10^6$ cycles at a frequency of 3.8 Hz under 200 N[7] loading with a stainless steel sphere-shaped piston (radius: 1.5 mm) at 37°C (ISO 6872)[x] (ER-11000, Technical and Scientific Equipment, ERIOS Ltd, São Paulo, Brazil).

AUT: Specimens were placed in autoclave at 134°C, under a pressure of 2 bar for 24 h.

AUT+M: Specimens were exposed to autoclave aging (AUT) followed by mechanical cycling (M) as described above.

X-Ray diffraction analysis

Quantitative analysis of phase transformation was conducted (n=2) in order to determine the relative amount of *m*-phase and depth of the transformed layer before and after aging procedures using X-ray diffraction analysis (XRD) (Model X'pert Powder, PANalytical, Almelo, The Netherlands) (Cu-Kα, λ=1, 54060 Åm, 45 kV, 40 mA, 25-80°, 0.02º angular pitch, integration time: 25 s). In mechanically aged groups (M and AUT+M), the analysis was performed on the surface where load was applied.

The relative amount (%) of transformed monoclinic zirconia on the treated surfaces was determined from the integral intensities of the monoclinic $(-111)_M$ and $(111)_M$, and the tetragonal $(101)_T$ peaks obtained after XRD.[28]

The transformed zone depth (TZD) (μm) was calculated on the treated zirconia surfaces where TZD was calculated from monoclinic phase relative amounts assuming that within the transformed surface layer, all tetragonal grains were transformed into monoclinic symmetry.[18]

Atomic force microscopy analysis

Atomic Force Microscope analysis (AFM) (Veeco Multimode, Nanoscope V, Plainview, NY, USA) was performed in the contact mode (10 µm x 10 µm) (n=2) in order to observe the surface morphology of the specimens used for XRD analysis.

Scanning electron microscopy (SEM)

One specimen from each group (n=1) was analyzed under Scanning Electron Microscopy **(**SEM) (Model INSPECT S50-FEI, Moravia, Czech Republic), under high vacuum at 5.000 magnification. This analysis was also performed to quantify the average grain size of a specimen from each control group.

Energy dispersive X-ray analysis

Morphological and chemical analysis was performed on the surface of control specimens from each group using SEM equipped with energy dispersive X-ray (EDX) device (Bruker, Spirit 1.0 software). The specimens were analyzed under SEM at high vacuum (20 kV, working distance: 12 mm) for 100 s.

Biaxial flexural strength test

The specimens were subjected to flexural loading (1 mm/min, 100 kgf load, in water) in universal testing device (Emic DL-1000, Emic, São José dos Pinhais, PR, Brazil) until catastrophic failure occurred (ISO 6872). The specimens were positioned with the treated surface (tensile side) down on the three supporting balls (Ø= 3.2 mm) 10 mm equidistant apart from each other in a triangular position. The assembly was immersed in water and circular tungsten sphere (Ø=1.6 mm) was used to apply load (1 mm/ min) until catastrophic failure. An adhesive tape (12 mm x 10 mm) (3M ESPE, Minn, St Paul, USA) was fixed on the compression side of the discs prior to the tests in order to avoid the fragments spreading and to provide better sphere-specimen contact.

Statistical analyses

The data were initially analyzed for homogenous distribution (Minitab software, version 16.01, 2010, Minitab Inc., State College, PA, USA). Since the results did not violate the homogeneity test, the mean and standard deviations of flexural strength data (MPa) of each group were analyzed using 2-way analysis of variance (ANOVA) and Tukey`s tests (3 levels: aging conditions: M, UAT, AUT+M) and 2 levels: zirconia types: (ZE and ZT). P values less than 0.05 were considered to be statistically significant in all tests.

Results

Biaxial flexural strength

Mean flexural strength was not significantly affected by the type of zirconia ($p=0.657$) but by the aging method ($p=0.000$) (Table 1). Interaction terms were not significant ($p=0.251$).

Regardless of the aging conditions, ZE (803.75) and ZT (815.61) did not show significant difference in flexural strength ($p>0.05$) (Table 2). Compared to the control group (C), aging with M only did not decrease the flexural strength significantly for both zirconia types ($p>0.05$) but aging with AUT, AUT+M significantly increased the mean flexural strength results compared to the groups C and M ($p<0.05$).

XRD analysis

Only AUT_{ZE}, AUT_{ZT}, $AUT+M_{ZE}$ and $AUT+M_{ZT}$ specimens showed monoclinic $(-111)_M$ and $(111)_M$ peaks, indicating $t \rightarrow m$ phase transformation (Table 3). TZD showed similar trends with the amount of *m*-phase being the highest observed for AUT_{ZE} (5.38 μm) compared to those of the other groups (6.18 - 5.38 μm). In other groups no TZD was observed (0 μm).

AFM analysis

Surface roughness (μm) was higher for AUT_{ZE} (0.09), AUT_{ZT} (0.08), $AUT+M_{ZE}$ (0.09 μm) and $AUT+M_{ZT}$ (0.09 μm) than those of other groups. The C groups for both ZE and ZT presented regular grains with visible outline (Figures 1a-b) but in M groups more granular surfaces were visible (Figures 1c-d). In the AUT and AUT+M groups, grain growth was evident with loss of defined grain margins also associated with small surface irregularities (Figures 1e-h).

SEM and EDX analysis

SEM images of C groups presented irregular surfaces with distinct boundary grains (Figures. 2a-b). Not in C (Figures. 2c-d) and M groups (Figures. 2e-f) but in AUT and AUT+M groups pieces of fractured grains at the surface were observed (Figures. 2g-h). C groups presented medium-sized grains for both ZE (0.636 nm) and for ZT (0.613 nm) specimens.

EDX analysis indicated the presence of O (20.5 w%) and Zr (79.42 w%) in ZE and O (21.9%) and Zr (70.02 w%) in ZT. Yttrium oxide could not be identified due to its low molecular weight.

Discussion

This study was undertaken to evaluate the biaxial flexural strength, surface roughness and transformation change in two types of zirconia after hydrothermal aging, mechanical fatigue and a combination of the both. Based on the results of this study, since the flexural strength results were not significantly changed after mechanical aging but increased after autoclave aging alone and with mechanical aging protocols but was not affected by the zirconia type, the first hypothesis could be partially accepted. As the amount of monoclinic phase increased after only autoclave and autoclave followed by mechanical aging, compared to the control groups, the second hypothesis could also be partially accepted.

In dental applications such as monolithic crowns, dental implants and abutments made of zirconia are exposed to saliva, temperature changes and cyclic loading during chewing that all may cause degradation in this ceramic and eventually decrease their mechanical stability.[2] In addition, when the veneering ceramic is removed during occlusal adjustments or fractured under occlusal loads, zirconia framework could also be exposed to the oral environment. Since long-term mechanical behaviour of FDPs made of zirconia is of clinical interest, evaluation of LTD could help predicting their clnical survival.

Typically transformation of zirconia crystals is activated and accelerated in the presence of water. In this regard autoclave vapour aging between 120˚C and 140˚C may effectively induce LTD and serve as a good aging method to study accelerated aging.[27] This type of hydrothermal aging coupled with repeated loading, simulating chewing function, could further accelerate degradation of zirconia.[2,29] Thus, degradation of zirconia at alternating temperatures through thermocycling[3] or autoclave cycling[2] are commonly used aging methods.[2,3,7,15] However, hydrothermal aging methods do not entail the mechanic component of aging which better translates for the clinical situation. For this reason, in this study, mechanical aging alone and in combination with autoclave aging were studied.

In the present study, as a consequence of aging protocols, the amount of transformation of *t*→*m* phase increased on the specimen surfaces but the level of *m* phase were not sufficient to cause decrease in the flexural strength of the zirconia materials studied. In previous similar studies, less aggressive aging protocols were employed, namely short duration of autoclave aging and less number of mechanical cycles with no drastic consequences on mechanical stability of zirconia.[1,3,15] According to the statistical analysis, the two zirconia materials tested presented statistically similar flexural strength which could be attributed to the similar microstructure, sintering procedures also being demonstrated by the EDX analyses.[3,27]

Interestingly, mechanical aging alone did not decrease the flexural strength values compared to the control group. One possible explanation for this result could be the low magnitude of load applied (200 N) that was not sufficient for *t→m* tranformation. In a previous study, mechanical cycling under a load that is 25% of the resistance of the material was not enough to decrease mechanical resistance[29] that also applied to this study.

Furthermore, the minimum TZD required to decrease zirconia resistance was reported to be between 11 to 14 µm.[2,3,17] Although in previous studies the number of cycles (10^6 and $5x10^6$) and load magnitude (100 N)[3] was less than that of this study, TZD (*t→m)* in this study was less than 11 µm. Similarly, despite the low magnitude of load (100 N) being distinct from this study (200 N), flexural strength values did not decrease.[16]

Interestingly, in this study AUT and AUT+M groups showed statistically higher flexural strength values compared to the C and M groups. This may be due to short period of exposure to AUT. Longer periods of autoclave exposure have been reported to cause significant TZD that decreased the mechanical resistance of zirconia.[3] The amount of monoclinic phase (66-71%) and TZD (5.38-6.18 µm) were consistent with those of the previous studies.[2,15] The increase in the phase transformation decreases residual stress and transformed zone volume inhibits crack propagation.[2] This volume increase might have led to compressive stress accumulation after LTD, that has possibly compensated for the tension stress caused by the mechanical testing. On the other hand, in another study autoclave aging (134ºC; 2 bar) for 200 h decreased the mechanical resistance of zirconia, resulted in 80% *t→m* transformation phase with TZD of 60 µm[9] where the duration of aging was considerably higher that that applied in this study (24 h). Similarly, in another study, 10 h of aging in autoclave at 125ºC increased the flexural strength but when the temperature was increased from 150ºC up to 225ºC, mechanical resistance decreased significantly.[15] Likewise, the authors reported 75% *t→m* transformation phase above 175ºC. Again in the present study, AUT was practiced at 134°C which is lower than this previous study.[15]

Mechanical aging alone was not detrimental for *t→m* transformation phase with TZD change. The mechanical cycling protocol was less aggressive than aging in autoclave alone or in combination with mechanical aging. In another study, a similar observation as made where mechanical cycling of $1x10^{-6}$ and $5x10^{-6}$ cycles, thermocycling (5-55ºC), water-storage for 200 days at 36ºC, autoclave aging (134ºC, 3 bar, 8 hours) and their combination was tested.[3] The authors reported no decrease in the flexural strength of zirconia and attributed this observation to the transformed area, which did not extend to the inner microstructure of zirconia.

LTD could be characterized with different methods.[4,5,6,14,17,20] In this study, the XRD was used in order to quantify the content of monoclinic and tetragonal phases before and after aging. When polishing was not performed after sintering, rougher surfaces were obtained which facilitate water penetration and favour phase transformation with LTD.[2] The fact that specimens were not polished after sintering may justify the high values of monoclinic phase (>65%) found in XRD analysis for both zirconia (ZE and ZT) after AUT and AUT+M. Moreover, after aging, the detachment of surface grains associated with volume increase in the grains was evident in SEM images after aging process.

The mean flexural strength values for both zirconia ceramics tested varied from 719.7 to 867.3 MPa for ZE and from 716.5 to 936.4 for ZT regardless of aging method. These values are slightly less than those of other studies,[13,21] but above the minimum flexural strength (500 MPa) required according to ISO 13356 and suffice the chewing forces in the molar region in the mouth.[1,15,22]

The phase transformation behaviour is directly related to the sintering temperature of zirconia.[6] When the ceramics are sintered above 1450ºC for 1 h, higher sensitivity to degradation under low temperatures has been reported.[12] Furthermore, the transformation rate may also be related to the size of grains in zirconia.[2] Since sintering protocols were not identical but similar for ZE (1530ºC) and ZT (1540ºC for 2 h), no significant differences were observed between the biaxial flexural strength of both ceramics, also resulting in similar grain size.

In an attempt to overcome chipping of the veneering ceramic, the indication of monolithic zirconia FDPs is increasing in reconstructive dentistry. Although mechanical properties were not affected by aging conditions in this study, it is crucial to monitor LTD of the zirconia ceramics tested due to increased $t \rightarrow m$ transformation phase after aging.

Conclusions

From this study, the following could be concluded:

1. Mechanical aging ($2x10^6$ cycles/3.8 Hz/200 N) did not decrease the biaxial flexural strength of both zirconia types tested.
2. The autoclave aging procedure (134°C, 2 bar for 24 h) alone or followed by mechanical aging, increased the flexural strength of the two zirconia types tested but also induced phase transformation from tetragonal to monoclinic and transformed zone depth compared to non-aged control groups.

3. SEM analysis indicated the presence of grain growth with loss of defined grain margins after both mechanical and autoclave aging.

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Captions to the tables and figures:

Tables

Table 1. Results of two-way analysis of variance (ANOVA) and the interaction terms for biaxial flexural strength (MPa) depending on the zirconia type and aging method ($*p < 0.05$).

Table 2. Mean biaxial flexural strength and standard deviations (MPa) for both ZE (E.max ZirCAD) and ZT (ICE Zirkon) zirconia types with and without aging conditions (C: Control; M: mechanical aging; AUT: Autoclave aging; AUT+M: Autaclave aging followed by mechanical aging). Different superscript letters in each column indicate significant differences (Tukey`s test, $*p < 0.05$)

Table 3. Amount of monoclinic phase (%) for both ZE (E.max ZirCAD) and ZT (ICE Zirkon) zirconia types with and without aging conditions (C: Control; M: mechanical aging; AUT: Autoclave aging; AUT+M: Autaclave aging followed by mechanical aging).

Figures

Figures 1a-h. Atomic Force Microscopy (AFM) images of zirconia ceramic (ZE, ZT) surfaces without and with aging: a) C_{ZE}, b) C_{ZT}; c) M_{ZE}; d) M_{ZT}; e) AUT_{ZE}; f) AUT_{ZT}; g) $AUT+M_{ZE}$; h) $AUT+M_{ZT}$. Note that C groups for both ZE and ZT presented regular grains with visible outline and M groups more granular surfaces. In AUT and AUT+M groups, grain growth was evident with loss of defined grain margins also associated with small surface irregularities.

Figures 2a-h. Scanning Electrone Microscopy (SEM) images (x5000) of zirconia ceramic (ZE, ZT) surfaces without and with aging: a) C_{ZE}, b) C_{ZT}; c) M_{ZE}; d) M_{ZT}; e) AUT_{ZE}; f) AUT_{ZT}; g) $AUT+M_{ZE}$; h) $AUT+M_{ZT}$. Note that C groups presented irregular surfaces with distinct boundary grains. In C and M groups grain deformation was not evident but in AUT and AUT+M groups pieces of fractured grains at the surface were observed.

Tables:

Effect	DF	SS	MS	F	*P*
Zirconia type	1	2816	2816	0.20	0.657
Aging	3	397428	397428	9.34	**0.000***
Zirconia*Aging	3	59355	59355	1.34	0.251
Error	72	1021188	1021188		
Total	79	1480788			

Table 1. Results of two-way analysis of variance (ANOVA) and the interaction terms for biaxial flexural strength (MPa) depending on the zirconia type and aging method (*$p < 0.05$).

Experimental Groups	ZE	ZT
C	779.9±114[a]	716.5±185.7[a]
M	719.7±130.8[a]	743.9±119.2[a]
AUT	848.1±75.9[b]	936.5±120.9[b]
AUT+M	867.3±49.3[b]	865.5±108.9[b]

Table 2. Mean biaxial flexural strength and standard deviations (MPa) for both ZE (E.max ZirCAD) and ZT (ICE Zirkon) zirconia types with and without aging conditions (C: Control; M: mechanical aging; AUT: Autoclave aging; AUT+M: Autaclave aging followed by mechanical aging). Different superscript letters in each column indicate significant differences (Tukey`s test, *$p < 0.05$)

Experimental Groups	ZE	ZT
C	0	0
M	0	0
AUT	71	66
AUT/M	71	66

Table 3. Amount of monoclinic phase (%) for both ZE (E.max ZirCAD) and ZT (ICE Zirkon) zirconia types with and without aging conditions (C: Control; M: mechanical aging; AUT: Autoclave aging; AUT+M: Autaclave aging followed by mechanical aging).